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IGNITION OF PROPELLANTS BY HOT GASES PART III

IGNITION ENERGY REQUIREMENTS AND HEAT TRANSFER UNDER TRANSIENT CONDITIONS

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20 OCTOBER 1955



U. S. NAVAL ORDNANCE LABORATORY

WHITE OAK, MARYLAND

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IGNITION OF PROPELLANTS BY HOT GASES
PART III. IGNITION ENERGY REQUIREMENTS
AND HEAT TRANSFER UNDER TRANSIENT CONDITIONS

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ABSTRACT: Ignition energy requirements were determined for propellants ignited in a time of 2-5 milliseconds by suddenly compressed inert gases. The twelve propellants tested included a wide range of compositions. Ignition energy values for the 2-5 millisecond range were found to be considerable smaller than ignition energies determined by others for longer ignition intervals.

Due to difficulties in determining absolute heat transfer to solids exposed to a gas whose mass flow, pressure and temperature vary simultaneously, ignition energy values quoted must not be taken as final. It is estimated that absolute energy values are good only to $\pm 35\%$; however, since many of the assumptions made cancel when various propellants are compared in the same apparatus, relative values should be good to $\pm 5\%$.

The results from these tests were combined with theoretical treatment and other workers' data for slower ignitions. It is concluded that the energy required to ignite a propellant is a function of the energy flux; at high flux rates less energy is required. Consequences of this conclusion are discussed.

It is therefore possible to systematize many of the variables encountered in ignition and to show that a simple thermal theory of ignition holds over the entire time interval range encountered in practical guns and rockets. Some applications of theory and data are discussed.

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This report covers progress on basic research on ignition mechanisms in solid propellants carried out under task NOL-B2d-02-1-55. Results should be applied only where there is a full understanding of the assumptions involved; they cannot be regarded as definitive and final. The opinions expressed are the responsibility of the author and the Fuels and Propellants Division of the Naval Ordnance Laboratory.

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IGNITION OF PROPELLANTS BY HOT GASES
PART III. IGNITION ENERGY REQUIREMENTS
AND HEAT TRANSFER UNDER TRANSIENT CONDITIONS

I. INTRODUCTION

The energy requirements for the ignition of propellants are of considerable interest from both theoretical and applied viewpoints. This report describes the determination of ignition energy requirements for propellants ignited in a time of the order of 2-5 milliseconds by hot, inert gases. The results from these tests are combined with theoretical treatment and other workers' data for slower ignitions. It is believed that the result is the most complete picture of ignition energy requirements which is presently available. In addition, some results on the transfer of energy to solid surfaces under transient exposures to hot high-pressure gases should be of interest in the heat transfer field.

Previous reports (1,2,3) must be referred to for a complete treatment of the locked-stroke compressor, recording apparatus, and previous results.

II APPARATUS AND INSTRUMENTATION

The locked-stroke compressor is designed to compress a gas very rapidly so that its temperature and pressure rise almost adiabatically. The final 80% of the pressure rise occurs in from 2 to 5 milliseconds, depending on the driving pressure and compression chamber pressure. The maximum point of the compression piston's movement can be mechanically varied. The useful range of compression ratios is approximately 4 to 30. The maximum allowable pressure is approximately 3000 psi. The maximum obtainable temperature varies with the gamma of the gas and is over 2000°K for monatomic gases. As the initial pressure and the compression ratio can be varied independently, the pressure and temperature can be varied separately, within limits. The compression piston is locked into position after only a slight backward movement.

A pressure-time trace is obtained through the use of an Aberdeen C-AN type pressure transducer, an amplifier and calibrator, an oscilloscope, and a drum camera.

Ignition is detected by a photomultiplier tube which "looks" through a window. The time of ignition is recorded by brightening the pressure-time trace.

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One modification has been added. A DuMont type 322-A double-beam oscilloscope has been used in place of the 304-H. This has allowed more precise recording.

III HEAT TRANSFER EXPERIMENTS AND RESULTS

A. Outline of Methods Used

As the methods used for determining heat transfer are novel, they will be described with some detail. The ideas and assumptions involved are simple. In order to prevent confusion by details, an outline of the ideas and assumptions involved is presented here. The heat transfer calculations which involve the most assumptions (those at the stagnation point of flow) are believed to be within $\pm 35\%$ of the correct absolute values and probably are better than this. Most errors are of the constant type and can be eliminated by the application of a constant factor to the data when more refined methods become available. The relative accuracy of ignition energy determinations for propellants is about $\pm 5\%$, though a propellant's variability may increase the spread in results to a figure greater than this.

1. It was experimentally proved that higher pressures obtained by compressing argon than with helium under identical initial pressures and compression ratios were due to the helium transferring more energy to the compressor walls than argon. This was done by measuring the effect of increasing the area exposed to the hot gases by introducing a flat coil (of round wire) with an area comparable to that of the compression chamber.

2. It was empirically determined from experimental data that the pressure-volume relations of each gas was regular enough so that adiabatic type thermodynamic equations could be used except that the adiabatic gamma (heat capacity at constant pressure/heat capacity at constant volume) was replaced by a constant (specific for the compressor used and different for each gas).

3. Equations for determining heat losses to the compressor walls were developed from the above empirical equations and thermodynamic laws.

4. To determine the heat lost per unit area of wall it was necessary to correct the area at the point of computation for the fact that some heat had been lost to the walls which were covered by the preceeding portion of the compression stroke. This correction was empirically determined by trial and error computation.

5. Large areas of copper, glass, and propellant-coated glass were separately introduced into the compression chamber. These were in the form of long cylinders (rods or wire) which had been

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made into flat spiral coils so as to fit within the chamber. These tests were compared with tests in which no coils were present. Differences in heat losses between loaded and unloaded chambers were due to the introduced coils except for three corrections: (a) Loaded chambers had larger cylindrical wall areas than the unloaded chambers at equal compression ratios, due to the effects of the volume occupied by the loading material on the final distances at which a given compression ratio was obtained. (b) Greater heat losses for the loaded chambers prevented the chamber walls from being exposed to the same temperatures present in unloaded chambers under equal initial and final pressure. (c) Competitive cooling by nearby surfaces was greater in the loaded runs (same volume of gas involved in heat conduction to more than one surface). Corrections for (a) and (b) were based on the area and temperature differences involved. Corrections for (c) were estimated from the differences between results for single coils and those obtained using two coils.

6. The above experiments showed that any differences between the heat transferred to solids of greatly different thermal properties were within the experimental error and thus small or absent. In other words, the properties of the gas are the controlling factor in heat transfer from a gas to a solid when the gas temperature is high and the time of transfer is short enough to prevent the rise in the solid's temperature from significantly decreasing the temperature difference between the solid and the gas.

7. It was thus possible to directly determine heat losses to any solid material in the plane of the face-plate or piston from parts 3 and 4 above.

8. To determine heat losses to a short cylinder suspended in the chamber, the no-flow calculations in 3 and 4 were multiplied by the ratio of flow to no-flow transfer found in 5 and corrections were included for the reduced inter-area competition for heat with the short cylinder as compared with a coil. This gave a value for an area uniformly distributed around the suspended cylinder.

9. In ignition experiments it was necessary to take into account the fact that the localized rate of heat transfer varied about the suspended cylinder (being greatest at the stagnation point). This correction was estimated from available data on the localized rate of transfer about a cylinder under steady-state gas flow, making some allowance for the fact that flow in the compressor was not steady-state.

10. A check of the accuracy involved in the measurements and corrections applied for changing from no-flow to flow conditions was made. This was done by igniting one of the propellants both in the plane of the face-plate (no-flow) and as a suspended cylinder (flow). The amount of energy per unit area required for ignition of the flat sample should be the same as that required at the stagnation point for the suspended cylinder. Within the precision

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set by the reproducibility of the propellant sample, the check was good.

11. Changes in heat transfer with speed of compression were roughly measured for two markedly different speeds. These measurements indicated an increase in heat transfer roughly proportional to the time of throw. As the apparatus is generally operated at the same compression speed, this will generally be the source of only a small error and no corrections have been applied for this. This type of error may become significant if two gases with different gammas (C_p/C_v) are being compared, or very large differences in pressures and temperatures exist between two determinations.

B. Early Experiments

During the course of some ignition tests, a long series of runs were performed under identical conditions (original pressure in compression chamber, compression ratio, and driving pressure) but with different gases, argon and helium. It was noted that the helium runs had a greater excess of driving pressure than the argon runs. The recorded peak pressures for the argon runs averaged 10.3% higher than for the helium.

According to the best data available on these two gases, they were still ideal in behavior over the pressures and temperature involved. This left the possibilities of leakage or of heat transfer to the walls. Shots that rebounded without locking had been tested and no evidence of leakage was found. It would also be unusual that leaks would show uniformity. The most likely explanation for the different results from the two gases seemed to be that they lost heat at different rates to the walls of the compression chamber.

To test whether heat transfer was the culprit, we inserted a flat coil of heavy copper wire in the compression chamber, with the plane of the coil parallel and near to the face-plate. The coil area was comparable to that of the chamber walls at the end of the compression stroke. It was reasoned that if heat transfer were the cause of the discrepancy, then the increase in area should reduce the pressure obtained with either gas, but should reduce the helium pressure the most. This is exactly what was found.

C. Measurement of Heat Transfer Based on Pressure Changes

Exploration of the possibility of using pressure drop as a basis for measuring heat loss was begun. By knowing only the initial pressure and volume and the pressure and volume at the

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maximum point of compression, the amount of heat lost could be defined to a value within fixed but farspread limits. The lower limit was the heat lost during a compression cycle which reached the final pressure and volume by first compressing isothermally and then adiabatically. The upper limit was the heat lost during a compression cycle which reached the final pressure and volume by first compressing adiabatically and then isothermally. Figure 1a illustrates these conditions. The area below line A is a measurement of the lower limit and that below B is a measurement of the upper limit, with the area between A and B being the amount of uncertainty (path dependent). In Fig. 1b it is assumed that the pressure and volume are known at one intermediate value. Clearly, such a point greatly reduces the area of uncertainty.

The volume and pressure at a number of points during the compression stroke were then determined. Three metallic probes were projected from the face plate at known distances of projection. Contact of the piston with these probes discharged a small capacitor so that a short, sharp, voltage pulse was applied to the Z-Axis of the oscilloscope. The pressure-time trace thus brightened at that point. Additional points were thus fixed at which it was possible to experimentally determine the volume and pressure simultaneously.

A plot of $\log V_0/V$ versus $\log P/P_0$ for the experimental values obtained gave a straight line. This result meant that the gases were obeying a regular type of law in which the adiabatic gamma (γ) was replaced by a non-adiabatic constant (unique for this compressor) which has been called alpha (α). The application of the method of least squares to these plots gave values of alpha equal to 1.562 for helium and 1.622 for argon. The Pearson correlation coefficient for both of these lines was 1.00. These plots are shown in Fig. 2 for argon and helium with heat loss and for an adiabatic compression. The scatter is large in the low-pressure region because a large part of the errors in pressure readings were constant in size (about ± 20 psi) so that they gave a much greater per cent effect at low pressures. If gamma is replaced by alpha in the usual equations for determining temperature, the temperature obtained will be an average temperature. The temperature of any central portion of gas which has behaved adiabatically can be obtained by using gamma and measured values of pressure in the pressure-based temperature equation. As an example, a central value of 2093°K and an average value of 1748°K was calculated for a helium compression. The alpha values found were tested by trying them on many other runs covering a wide range of compression ratios and initial pressures. These showed rather convincingly that a good prediction of pressure could be obtained by using experimentally determined alphas. It was thus possible to do much better than determine pressures and volumes simultaneously at a few points. They could be calculated at any point.

This made it possible to develop an equation for calculating the heat lost up to or between any desired intervals of the compression

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stroke. Rates of transfer can also be derived from this data. This equation was derived from a basic thermodynamic equation in the following manner:

(1) Starting with the familiar -

$$dQ = C_p dT - V dP, \text{ where}$$

Q = heat, C_p = heat capacity at constant pressure,
T = temperature, V = volume, and P = pressure.

(2) The value of dP is found in terms of the empirical equation involving a constant alpha -

$$P = P_o (V_o/V)^\alpha$$

$$dP = -\alpha P_o V_o^\alpha dV/V^{\alpha+1}, \text{ where subscript "o" indicates initial state.}$$

(3) Substituting this into the original equation and integrating

$$\Delta Q = C_p(T_f - T_o) + \frac{\alpha}{1-\alpha} P_o V_o \left[(V_o/V_f)^{\alpha-1} - 1 \right]$$

is obtained, where "f" indicates final state and ΔQ is the heat lost.

(4) Substituting, using the equation

$$T_f = T_o (V/V_o)^{\alpha-1}, \text{ and simplifying -}$$

$$\Delta Q = (C_p T_o + \frac{\alpha}{1-\alpha} P_o V_o) \left[(V_o/V_f)^{\alpha-1} - 1 \right]$$

$$= (C_v T_o - \frac{1}{\alpha-1} P_o V_o) \left[(V_o/V_f)^{\alpha-1} - 1 \right]$$

(5) By using the relationship $PV = (C_p - C_v)T$ we obtain -

$$\begin{aligned} \Delta Q &= - \frac{P_o V_o (\gamma - \alpha)}{(\gamma - 1)(\alpha - 1)} \left[(V_o/V_f)^{\alpha-1} - 1 \right] \\ &= - \frac{P_o V_o (\gamma - \alpha)}{(\gamma - 1)(\alpha - 1)} \left[(P_f/P_o)^{\frac{\alpha-1}{\alpha}} - 1 \right] \end{aligned}$$

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The last two forms of this equation are particularly convenient for calculation purposes. With them the amount of heat lost to the compression chamber walls can be easily calculated at any moment in the compression stroke.

Although the alphas gave a good prediction of the pressures experimentally found, there is reason to doubt their constancy at low compression ratios. The area exposed to the hot gases can be separated into a fixed portion (the end area of the piston plus the equal area of the face-plate) and a variable portion (the cylindrical walls). The area at the beginning of the stroke is considerably larger than that at the end of the stroke. This may be partially counterbalanced by the much lower temperature gradient at the beginning of the stroke than at the end. Our principal interest in these alphas is with respect to their effect on the amount of heat transfer. Examination of the bracketed term in the above equation (plus checks by actual computation) shows that the amount of heat transferred is small during the low compression region during which the principal change in area occurs. (Only 17 per cent of the heat loss would occur while an initial 54 per cent of the change in wall area was occurring for a peak compression ratio of 15 and using helium.) In other words, the major part of the heat is transferred while the wall area shows only a modest change. In summation, it seems possible that alphas may not be strictly constant for low compression ratios, but this probably has only a slight effect on most of the heat transfer calculations.

To translate results for the total heat loss to the chamber walls to the heat loss per unit area of exposed wall at some point during the stroke, some correction must be made for heat lost to the walls which have been covered at that point. For a slight movement of the piston the area would be the final exposed area plus $1/2$ the area covered by the piston movement. This formula gives too large an area when the length of the stroke becomes appreciable, because the rate of transfer per unit of movement increases as the piston moves forward. By trial and error it was found that if the covered area was divided by twice the square root of the compression ratio and this was added to the final area, a close approximation to the correct area was obtained. By dividing the total heat loss by this value, the amount which the final walls received per unit area (final referring to the point of interest) was obtained.

It was thus possible to calculate the amount of heat lost to each unit of exposed wall area at any point during the compression.

D. Heat Losses to Coils of Various Materials Suspended in the Compression Chamber

These experiments were designed to determine how large an effect the thermal properties of a solid had on the amount of heat

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transferred to it under a transient exposure to hot, high-pressure gases and to obtain data on which heat transfer calculations to short cylinders of propellants could be based. To obtain significant pressure differences the determination of heat losses was made on long cylinders made into a flat coil. The materials tested and their thermal diffusivities in c.g.s. units were as follows: copper, 1.14; pyrex glass .0058; and propellant (principally nitrocellulose, coated on pyrex glass) .0008.

Copper and pyrex glass were tested in the form of long thin cylinders (wires and rods) coiled into flat spirals whose outer turns were nearly the diameter of the compression cylinder. The copper wire was .102" dia. and long enough to provide an area of 99.4 cm². The pyrex rod was .107" dia. and provided an area of 111.6 cm². The areas of these coils were comparable to those of the compression chamber walls as can be seen by the comparable heat losses tabulated in Table I. Three runs were made for each material and for a blank; the runs being alternated. Adjustments were made for the volumes occupied by the glass and copper coils so that the trials were made under approximately equal compression ratios. The effects of any slight differences in compression ratios were eliminated by determining an alpha for each run and then averaging for the group. These average alphas were used in the above developed heat transfer equations to determine the total amount of energy lost to each system under conditions of equal final pressures and equal starting conditions for the three systems. These values are given in Table I. (Note that comparisons under equal compression ratios give similar results but do not give as close a comparison because peak pressures and temperatures would be different for different alphas). The blank values were corrected for the fact that these calculations were for higher gas temperatures than the walls of the loaded systems were exposed to. The bracketed term in equation 5 is equal to the temperature difference between the gas and the walls divided by the initial temperature. The blank values were reduced by multiplying them by the ratio of the value of the bracketed term for the loaded system to that for the blank system. A second correction was applied to increase the value for the blank system to correct for the fact that the average cylinder wall area exposed was smaller than for the copper and glass coil systems. This was true because of adjustments necessitated by the volume occupied by the glass or copper. These two corrections were applied so that the amount of energy lost to the walls under the conditions and areas holding for the glass and copper systems could be accurately determined. These corrections, though significant, were not large as can be seen from Table I where corrected (column 6) and uncorrected (column 5) blank values are shown. There was some wall to coil competition for heat in the loaded system. Omission of a correction for this has the effect of assigning a greater heat loss to the walls of the loaded system than actually occurred. Similar effects are discussed further in this section, and an estimated correction has been used in applying this data to short cylinders. By subtracting the values in column 6 of Table I from the overall values for the

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loaded system; the heat lost to the coils alone was obtained. Division of these values by the respective areas of the coils gave the heat loss per unit area of coil. This was an average value for an area equally distributed around the rod's circumference.

A propellant was tested in the form of a 15 to 20 micron thick coating on glass coil supports. This coating was principally nitrocellulose. It was prepared by dipping the coils in a solution of IHBE-152 propellant (96.16% NC (12.6N), .96% diphenylamine, 2.88% K_2SO_4 , and 4.77% total volatiles) in ethyl acetate and allowing to dry; repeating the process until an adequately thick coating was obtained. To prevent webbing between turns of a coil, it was necessary to use coils with a greater spacing between turns than for the above runs. So that a large cooling surface could still be maintained, two coils were mounted parallel to each other and with a spacing of about .10" between coils. This gave less cooling per unit area of coil than the single coil arrangement, due to the crowding causing an increase in competition for heat by adjacent areas. The error introduced by not correcting the transfer to the cylinder walls for coil to wall competition is also increased; making the values found for the coils appear to be appreciably less than they are. By making tests with the same glass coils, bare and coated, an excellent comparison of heat transfer between glass and propellant could be obtained. As good single coil data for pyrex glass had already been obtained, single coil data could be indirectly obtained for the propellant coated glass.

Heat transfer values for these dual coil experiments are also given in Table I, using the same type of calculations as for the single-coil data. As expected, double-coil values were much smaller than single-coil values. Differences between the two materials were small, but were observed with both argon and helium as operating gas. The propellant coated coils actually gave slightly larger values than did the glass. These differences are probably within the errors involved in the measurements. Such a difference would be in opposition to thermal property origin, but a greater degree of surface roughness for the propellant might be a plausible explanation.

These experiments showed that about the same amount of heat was transferred to surfaces of copper, glass, and propellant. Greater crowding in the double-coil experiments resulted in a lower heat transfer per unit area than for the single-coil experiments. This effect was large enough so that an adjustment for the reduced surface to surface competition for heat present for a short cylinder compared to a single coil was applied in the following section.

Apparently, the difference in temperature between the gas and solid is the determining factor in heat transfer from a given gas. The fact that the amounts and rates of heat transfer to materials differing by a factor of as much as 1400 in thermal

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properties were almost identical should be generally true when the increase in surface temperature is only a small fraction of the temperature difference between the solid surface and gas. This condition is met under the short times of application and high gradients existing during the compression stroke of the apparatus. Here there are temperature differences of the order of one to two thousand degrees Kelvin and surface temperature rises varying from a few degrees for the copper to a few hundred degrees for the propellant. It should be noted, however, that the surface temperatures reached by solid surfaces under equal heat transfer rates are quite dependent on thermal properties of the solid.

The suspension of the coils within the chamber exposed them to gas flow, varying during the stroke from 0 to maximum values of the order of .5 mole/cm² sec. The values of heat transferred per unit area for the single coils were thus larger than for unit area of walls, as there is no flow involved in transfer to the piston end or to the face plate. Values of this flow/non-flow ratio were needed for calculations below. As all the materials behaved similarly, the values for glass were used because less scatter was present in these data. Ratios were 1.05 for helium and 1.93 for argon.

K. Heat Transfer at the Stagnation Point of Short Cylinders

Inhibited-end cylinders of propellant were used as samples in the tests on which ignition energy determinations were based. This shape may be considered as a short form of the above coil. We would thus expect that a similar amount of energy would be transferred per unit area of cylinder as per unit area of coil. To apply heat transfer data from coils to the calculation of transfer to cylinders, adjustments were made for the non-existence of intra-coil and negligible cylinder to wall competition for heat. Ignition would be expected to occur at the point of maximum heat transfer (stagnation point), so overall values were changed to localized values for the stagnation point regions. We were thus able to base heat loss calculations on pressure differences resulting from large area changes and apply the results to cases where pressure differences would have been too small to measure. The determination of the heat transferred per unit area of wall at any point during the compression stroke has already been discussed. These calculations were utilized by expressing the results in terms of the ratio of the transfer per unit area at the stagnation point to that at the walls.

The single-coil values were arbitrarily increased by 20% to obtain values for short cylinders where there is little inter-surface competition for heat. This estimate was based heavily on the differences in energy transfer values found between the double-coil and single-coil experiments. Such values are overall values for areas distributed uniformly about the circumference of the cylinder. It has been shown (4,5) that, under steady state conditions for a cylinder suspended in flowing gases with its longitudinal axis normal to the direction of flow, the maximum ratio of the heat transfer rate at the stagnation point to the

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overall value is 1.7 and for low flow rates is slightly less. The direction of flow about a cylinder of propellant mounted in our compressor with its longitudinal axis parallel to the face plate meets the above conditions. The flow rate is definitely not steady state, however, differing in the following ways: 1. The flow rate continuously changes during the compression stroke. 2. It increases with the distance of the cylinder surface from the face-plate. 3. The surface layers about the cylinder (and all other layers) are continuously being heated during the compression and also increasing in pressure and density. We have definitely seen evidence, on samples which were discolored or partially decomposed but did not burn, that there was more discoloration and decomposition (thus heat transfer) at the stagnation position. There was weak evidence of higher than average heat transfer at a position diametrically opposite the stagnation point, which is also in agreement with the higher than average transfer predicted there under steady state flow (5). With the above as a basis, an arbitrary factor of 1.5 was selected to change overall values to values at the stagnation point. A qualitatively similar factor was inferred by noting the increase in conditions needed to cause sample darkening to spread from the stagnation point until the entire cylinder was darkened for some samples.

If we apply these two corrections to the above-given values for single-coils, we obtain ratios of 1.89 for helium and 3.47 for argon. These values are for the amounts or rates transferred at the stagnation point of a suspended cylinder versus the amount or rates transferred at the walls.

As the correction factors used above were given rather arbitrary values, an attempt was made to experimentally verify them. Arcite 141 was the only propellant which seemed likely to receive enough energy to ignite as a flat surface on the face-plate. A disc of Arcite 141 was imbedded in a disc of modelling clay which partially covered the window in our compressor, with the clay slightly overlapping the disc edge. Only a flat surface was thus exposed to the gas. This was approximately in the plane of the face-plate so that flow was negligible. If the factors were correct, then the same values of ignition energy should have been found with the suspended cylinders using those factors, as were found for the flat surfaces using a factor of unity. For helium a value of .121 cal/cm² was obtained for flat surfaces and .117 cal/cm² for cylinders. For argon a value of .086 cal/cm² was obtained for flat surfaces and .116 cal/cm² for cylinders. This is a good check considering the scatter in results due to inhomogeneity of propellant. The checks are by no means perfect, however, and we are trying to further refine our data by the direct measurement of surface temperature with a thin thermocouple.

F. Changes in Heat Transfer with Speed of Compression

To test this effect helium gas was used, the exposed area was increased with a copper coil, and driving pressures were alternated between 220 psi and 280 psi. The compression speeds alternated from 3.7 milliseconds to 2.7 milliseconds for the final 80% of the

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pressure rise, the faster throw being for the higher driving pressure. The amounts of heat transferred per unit area of the composite system changed from .114 to .085 cal/cm², the greater quantity being for the slower compression speed. We thus found that there was a 25% decrease in the amount of heat transferred when the speed was increased by 36%. The rate of transfer for the faster throw was only 2% higher than for the slower throw. The amount of heat transfer is thus closely proportional to the time of throw for the two points measured.

This change in the amount of heat transfer with compression time is undoubtedly the source of some of our scatter. Ordinarily only a slight excess of driving pressure is used over that required to lock the piston. Tests will thus ordinarily be made at similar driving speeds. Runs which are made so that large differences in compression ratios are obtained or in which gases of different gammas are used will show appreciable differences in driving speed. This corroborates some observations made in an earlier report (2, p.9), in which some discrepancies were attributed to differences in driving speeds.

G. Further Discussion of Heat Transfer

In earlier reports (2,6) an hypothesis was advanced for the calculation of heat transfer to a solid from a gas. This hypothesis was based on the frequency of collision of the gas molecules with the solid surface, their average energy, and a proportionality factor. Some ignition data indicated that the proportionality factor varied with the gas. The possibility of this being due to the differences in accommodation coefficients arising from different molecular weights and for diatomic gases the possibility of different accommodation coefficients operating on rotational energy exchange was mentioned. The present work indicates that this discrepancy in proportionality factor was due to differences in molecular weights but was purely a flow effect for monatomic gases. From Table I it can be seen that for transfer to the wall surfaces the ratio of the amount of heat transferred by helium to that transferred by argon is 2.8. This compares favorably with the value of 3.2 predicted by the hypothesis and assuming a constant proportionality factor. For transfer to the coils the values in Table I give a ratio of 1.5, showing clearly that the effects of flow enhance the effectiveness of heat transfer of argon more than for helium. The ignition data was from samples which were mounted so that flow was definitely a factor. Although we have investigated the heat transfer properties of nitrogen, we intend to refine our work before drawing conclusions. One complication present in nitrogen calculations is that this gas is imperfect enough over the temperatures and pressures covered to show changes in gamma sufficient to cause significant effects in heat transfer calculations.

The alphas which we used in our heat transfer equation for calculating heat losses to the walls are, of course, directly

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applicable only to our particular compressor system. It would be quite useful if we could calculate heat losses for other compression systems. By making the assumption that for a given gas under equal starting pressures and temperatures and the same compression ratios the same amount of heat will be lost per unit area of wall, regardless of the system, we can do this. Then we only need to calculate the amount of heat lost per unit area of wall for our system and multiply this by the area of the other system's walls at the point of comparison. Alternatively, we could make this assumption for a given set of conditions and work backward to obtain an alpha for the other system. This new alpha could then be used over a large range of conditions. Some correction for different speeds of compression could be applied from the above data on this. Possibly, differences arising from different length to diameter ratios may exist. We hope to experimentally determine how much the heat transfer at the cylindrical walls (where a peculiar type of flow is involved) differs from that at the piston end and face-plate.

IV THE DETERMINATION OF IGNITION ENERGIES
OF A SERIES OF PROPELLANTS

These determinations simply amounted to the application of the heat transfer results to ignition experiments given in an earlier report (3). The compositions of the propellant samples and their preparation, mounting, and testing are described in that report. A few recent tests with flat samples of Arcite 141 have already been mentioned, but otherwise the ignition energy values are based on experiments in which cylindrical samples with inhibited ends were used.

Ignition energies were determined from tests in which the minimum conditions necessary for ignition were involved. These ignitions may occur at, before, or after the peak pressure and temperature for a given test, but the important heat transfer period occurs before the peak. The statistical nature of ignition is discussed in reference 3. If we made a number of tests under the same minimum conditions, some of these would ignite at the peak. These ignition energies are thus considered to apply for ignition processes in which the energy is transferred in three milliseconds (during which the final 80% of the pressure rise occurs). If we want the probability of ignition occurring at or before the peak to be high, a small increase in the minimum energy for three millisecond ignition (of the order of 10%) will do this.

Table 2 lists the results of our ignition energy determinations on propellant cylinders with inhibited ends and using helium and argon as gases. Average values and average values increased by ten per cent are also listed. Average values range between .12

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cal/cm² for Arcite 141 and greater than .33 for EX-7077. These ignition energy values are much smaller than those given by other investigators (7,8,9), but their work was carried out under conditions giving much longer ignition times. The argon values average 18.5% lower than the helium values. This difference probably is a result of constant errors in our heat transfer calculations and can be corrected by application of a constant factor upon refinement of measurement. The average values are probably better values than those for either gas, but errors were probably greater in the argon heat transfer determinations. (The coil experiments yielded smaller values of heat transfer with argon than with helium; thus, errors of fixed magnitude loomed larger with respect to the argon results.) In view of the smallness of the discrepancy, the results are in good agreement with our earlier expressed ideas (2,6) that ignition energies should be independent of the donor gas.

The propellants in Table II are arranged in the order of their relative ignitabilities reported in reference 3. For inhibited-end cylinders that arrangement was based on the lowest peak pressures and temperatures which would bring about ignition, the highest peak conditions which failed to bring about ignition, and the values of peak conditions at which ignitions occurred but burning did not propagate (for some samples). Values of ignition lags for many propellants tested in the form of thin, stacked discs were also included in arriving at this order. The ignition energy values tabulated in Table II were derived only from the minimum peak conditions which would bring about ignition for inhibited-end cylinders. The order is essentially unchanged from the order derived from the relative ignitability tests. This correlation is clearer if comparison is made from a graphic chart in the previous report (3), in which the degree of overlap between samples is shown. Experimental scatter plus poor homogeneity of samples is probably responsible for some of the variations present, particularly for OGR.

The correlation of ignition energies with chemical composition is parallel to the earlier discussion of the correlation of relative ignitabilities with chemical composition (3). Some recent tests with explosives have yielded similar ignition energies to propellants of similar chemical structure. These results will be given in a later report, after a wider range of explosives have been tested.

V CORRELATION OF THESE AND OTHER DATA WITH THEORY

A. The Reduction of Ignition Energy Requirements by an Increase in Energy Flux

Lewis and von Elbe (7), Shook (8), and Enig (9) have reported ignition energy values. Enig's calculations cover a range of ignitions occurring in from 150 milliseconds to 30 seconds, being

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based on data from the ignition of propellants by hot gases by Brier, Churchill, Thatcher, and Engibous for forced convection (5,10,11) and Bickford, Conant, Wachtell, and Roth (4,12) for natural convection. Those experiments also included a variety of gases and conditions. The propellants we have tested do not include M-2 propellant, but its composition is intermediate between that of JPN and IHBE-152. We have thus estimated its ignition energy as being an average of those two propellants and equalling .25 cal/cm² for the 10% above average value, which corresponds to a high ignition probability after a 3 millisecond time of heat transfer. Enig showed that the University of Michigan and Franklin Institute experiments were carried out under such conditions that the maximum and minimum flux were quite close. Constant flux can thus be assumed and, by dividing his ignition energies by the ignition times, the flux for each test can be obtained. For our data an average flux was obtained by dividing the ignition energy by three milliseconds. Note that this is an average flux and not a constant flux. It was thus possible to obtain ignition energy versus energy flux for a range of ignition times varying from 3 milliseconds to 30 seconds.

The above relationship is plotted in Fig. 3 for M-2 propellant. The log-log plot gives a straight line relationship over the entire range and the line can be reasonably extrapolated to a high flux region which would give ignitions in a time of 100 microseconds. There is thus a regular decrease in ignition energy requirements as the flux rate increases for the entire range of ignition intervals encountered in rockets and guns.

Figure 4 is a log-log plot of ignition energy versus time of ignition. Figure 5 is a log-log plot of energy flux versus time of ignition. Figure 6 is a log-log plot of ignition temperature versus time of ignition. These graphs give useful straight line relationships, but care must be taken not to draw conclusions from them which are in conflict with Fig. 3. The ignition temperature data was taken from Franklin Institute calculations (4,11) on University of Michigan and Franklin Institute data.

B. A Thermal Theory of Ignition and a Graphic Model for Applying this Theory.

The data in Fig. 3 plus the Franklin Institute data on ignition temperatures provide strong proof that a simple thermal theory of ignition holds quantitatively over the entire ignition range encountered in guns and rockets. This theory simply assumes that ignition of a propellant occurs when the external source of energy increases the propellant's temperature to the point that the decomposition rate is sufficient to cause self-supporting burning. The rate of energy transfer, duration of transfer, and thermal properties of the propellant determine the surface temperature history and gradient within the propellant. The chemical properties of the propellant, of course, determine the effect of temperature on the decomposition and the overall rate necessary to produce enough heat to support

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combustion. As less energy is required to reach a given surface temperature when the energy flux is increased, less energy is needed for ignition as the flux is increased. This is partially counterbalanced by the fact that higher surface temperatures must be obtained to support combustion when the thermal gradient is increased by an increase in flux. (The thickness of the zone in which decomposition is occurring decreases and there is also a higher sub-surface rate of thermal diffusion.) Ignitions may occur in which the role of the external source is reduced, but these will be longer than those considered satisfactory for rocket or gun ignitions, or, will be unsatisfactory in other ways, such as irregularity or by a dangerous accumulation of incompletely burned products.

A graphic model based on the above theory is shown in Fig. 7. This model is constructed by first making a log-log plot of the increase in surface temperature versus the time of energy transfer. For a constant energy flux this surface temperature can be determined by the application of the equation shown in Fig. 7. (taken from Carslaw and Jaeger (13)) to a material of known thermal diffusivity and thermal conductivity. The thermal properties used in Fig. 7 were taken from Shook's determination for JPN propellant (8). Judging from Rainbird and Ward's data (14), these are probably quite close to those for M-2 propellant. Once the simple calculation of the increase in surface temperature versus time has been made for a given flux, a family of curves covering a wide range of fluxes can be constructed as shown. As the product of the flux and time is equal to the energy input, it is also quite simple to construct a family of constant-energy lines, as shown. Up to this point no restriction has been placed on the original ambient temperature, except that the thermal properties of the propellant must be applicable over the range covered. To plot a definite value of temperature on this graph, a value must be assigned to the ambient temperature. The ignition temperature versus time line taken from Fig. 6 is plotted for three different ambient temperatures.

The use of this graph can now be illustrated. If either the ignition energy, energy flux, ignition time, or ignition temperature, is known, values of the other three can be immediately obtained. These variables are simply read where the known variable intersects the ignition temperature line.

The ignition temperature line was drawn for three different ambient temperatures to show the change in ignition energy requirements with changes in the ambient temperature of the propellant. (Note that these lines are straight but diverge.) It can be easily seen that at an ambient temperature of 236°K (-35°F) and a flux of 160 cal/cm² sec., 1.6 times as much time and energy are needed as at 322°K (120°F). Similarly, at a flux of 5 cal/cm² sec. a factor

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of 1.9 is involved. This large change in ignition energy requirements with ambient temperature is one of the important factors responsible for the large changes in ignition behavior with changes in the ambient temperature of the propellant.

The power of this model may be better realized by noting that it contains all the information in the previous four graphs plus much other information. We have been using it for several years as a basis for testing the thermal theory of ignition. This model places stress on the rate of energy delivery rather than the age of delivering it. Although our data is from hot gases, the model should be generally applicable where the heat is transferred by a conduction or convection process. This includes hot gases, hot solids, hot liquids, heat released from a phase change of the ignition products, and heat released from the recombination of dissociated molecules. The energy required for ignition by radiation would be expected to be larger as the absorption does not all occur at the surface; thus equal energy fluxes and times would lead to a lower surface temperature than if absorption were all at the surface plane. This is borne out by Shook's work (8) with JPN propellant. His ignition energies from a radiant source are larger than would be expected for transfer by a convection or conduction process. Courtney-Pratt and Rogers have shown this quite clearly by recent work (15). They showed that the initiation energy (by radiation) required for silver azide can be markedly decreased by increasing the absorption coefficient by the addition of a dye. (This energy was also quite similar to the extrapolated value we would get for M-2 propellant at the less than 61 microsecond time of heat transfer involved.) It is conceivable that energy arising from chemical or catalytic action of the ignition system products with the propellant surface might be treated as just another thermal source; however special assumptions would have to be made, such as considering the attacked layer of the propellant surface as part of the ignition energy source and the surface beneath this as being the ignited surface.

Thermal properties, particularly thermal diffusivities and conductivities, may be very important in determining differences in ignition energies and relative ignitabilities of propellant. Clearly it would be expected that a material with a high thermal diffusivity and conductivity would acquire a much lower surface temperature than one of much lower thermal diffusivity and conductivity under equal conditions of rapid heating. Thermal properties can conceivably have a large range for propellants. The determination of the actual variations is important. The effects of metallic additives, such as the incorporation of fine wires, or of inorganic salts in even small quantities might be more important than realized. (The values of thermal diffusivities for some typical materials in c.g.s. units are 1.14 for copper, .0058 for pyrex glass, and .00085 for JPN propellant.) If only more thermal data were available, the ignition energies in Table II could be used to assess the relative importance of thermal properties to chemical properties in determining these values. Thermal

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properties of propellants have been reported by Shook (8), Altman (16), Ward and Nicholson (17), Rainbird and Ward (14), Thiokol Chemical Corporation (18), and Atlantic Research Corp., (19). Rainbird and Ward's data are sufficient to show one striking fact. The thermal conductivity of the only high nitroguanidine composition which they tested was more than fifty per cent greater than for any one of a series of S.C. and related cordites (double-base composition consisting principally of nitrocellulose and nitroglycerin). These cordites varied by only about twenty per cent among themselves. This is directly in agreement with the much higher values of ignition energies found for propellants containing large quantities of nitroguanidine than for ordinary double and single base propellants. The poor ignitabilities of cool picrites may thus be a direct result of their different thermal properties. If we accept this, then improvement of their ignitabilities by reducing the accumulation of "fizz" products must be simply considered as one method of alleviating a condition which has its roots in thermal properties which necessitate a greater transfer of energy for a given temperature rise than is required by ordinary double and single base propellants.

The above thermal theory is quite similar to the thermal theories of Lewis and von Elbe (7) and Frazer and Hicks (20,21), but differs in the following aspects: 1. There is no indication of the existence of a minimum ignition energy over the entire ignition time range of guns and rockets, so the hypothesis of Lewis and von Elbe that ignition energies will approach a constant value is omitted from our theory. It is conceivable that such an absolute minimum value may be reached under conditions sufficient to cause ignitions in a few microseconds or less. From 100 microseconds on up to much larger times of ignition the concept of a minimum ignition energy seems only to have meaning if we relate it to some definite value of energy flux. 2. We have focused attention on the rate of energy transfer rather than the transfer coefficient. We consider the transfer coefficient as being only one of the variables involved in determining the rate. This simplification permitted the construction of our graphic model. 3. We have explicitly stated that the surface temperature necessary for ignition must increase as the energy transfer rate increases and have given reasons for this. Previous workers have been vague on this point. Hicks and Frazer stated that if the ignition temperature is assumed to be a constant then a graph such as Fig. 3 should give a slope of one. Actually, the slope is 0.79, in line with the requirement of a higher temperature being required for ignition as the transfer rate is increased. This increase in temperature requirements is also supported directly by the data in Fig. 6, but the data points do not cover as wide a range as in Fig. 2. The assumption of a constant ignition temperature will thus lead to low results in ignition energy calculations, but is still a useful approximation in the absence of data on ignition temperatures taken at several rates of transfer.

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C. Some Applications of Theory and Data

Our data covers ignition energy data for a series of twelve propellants at a three millisecond ignition time. Ignition data is presently available over a wide range of ignition times for only M-2 propellant, although some experimental data is yet unanalyzed. More data should become available by the continuance of present programs such as those at the University of Michigan, Franklin Institute, the Bureau of Mines (22), Chattanooga University (23), and NOL. Although extension of ignition and thermal property data will greatly expand and simplify the application of ignition energy data to practical problems, we already have enough information to begin such work. Our data is for an ignition time which is comparable to that of many guns and some rockets. It can be seen by reference to M-2 that ignition energies change slowly enough with changes in flux to permit a modest extrapolation of our data. In this way we can cover the major portion of the range of ignition times occurring in guns and rockets. The series of propellants that we have tested covers a fairly wide range of propellants and can be extended by interpolation to cover many closely related propellants. We might thus begin to look for practical comparisons and applications.

It would seem that if the area of a propellant were multiplied by its ignition energy requirements per unit area under fluxes approximating those in practical systems, a quantity approximating the ignition energy requirements of a propellant system would be obtained. We are presently trying to determine how dominant this factor is with respect to other variables for existing gun and rocket systems. The comparisons we have so far made have shown a surprisingly close correlation for guns, but have indicated that rockets require considerably more than this quantity (less efficient utilization of energy from the ignition source).

We have already pointed out the large changes in ignition energy requirements with the ambient temperature of the propellant. A quantitative analysis of this effect for a wide range of propellants will require more thermal and ignition temperature data. It is clear from Fig. 7, however, that this change will be large for all propellants. Thus, this brings out the important fact that ignition systems which deliver more energy (or deliver it more rapidly) as the temperature is decreased are badly needed. For most ignition systems the reverse is probably true, though McLennan (24) has reported a primer with a negative temperature coefficient. The development of primers which adjust their energy delivery rate to match the requirements of the propellant over a wide temperature range should be an important step in obtaining satisfactory

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ignition over a wide temperature range. It is also obvious that the temperature at which the highest rate of transfer is needed will sometimes coincide with the point at which embrittlement of the propellant has decreased its resistance to physical shock. Great care must thus be taken in such negative coefficient systems to ameliorate shock as far as possible.

It has already been mentioned that the thermal properties of a propellant which determine the amount of energy required to reach a given surface temperature under a given flux may be as important as the chemical properties which determine the temperature necessary for ignition. It is hoped that thermal data will become available for all propellants or at least a much larger range of propellants. This should permit a clearer assessment of the relative importance of physical and chemical properties in determining ignition energy requirements.

VI SUMMARY

1. Methods have been developed which permit the evaluation of amounts and rates of energy transfer to the walls of a compression chamber and to objects suspended within it.

2. These measurements showed that the rates (and amounts under equal times) of heat transfer were independent of the nature of the solid surface under the high gradients and short times of transfer existing during a compression occurring in a few milliseconds. This independence should be general whenever the rise of the surface temperature of the solid is small with respect to the difference in temperature between the gas and the solid during the time of transfer.

3. The ignition energies of twelve propellants under conditions adequate to cause ignition in three milliseconds have been determined by applying the above type heat transfer calculations to propellants ignited in our compression apparatus. These values were all much smaller than ignition energies found by others under conditions which gave much slower ignitions.

4. By combining some of the above data with other data for slower ignitions we have been able to show that ignition energies and times continuously and regularly decrease as the rate of heat transfer is increased. This relationship holds for ignition intervals ranging from 30 seconds to 3 milliseconds based on experimental data and can be reasonably extrapolated to 100 microsecond intervals. This covers the entire range encountered in gun and rocket ignitions.

5. A thermal theory of ignition is presented which should be applicable to a wide range of ignition problems. A graphic model embodying this theory is given. This can be very useful

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for such purposes as calculating ignition energies and times under various rates of energy transfer and ambient temperatures of the propellant.

6. The ignition energy data and theory clearly bring out the practical importance of certain factors. The influence of the product of the area of a propellant and its ignition energy per unit area in determining the amount of energy which practical ignition systems need to supply should be investigated. This shows some promise of forming a scientific basis for determining the amounts of energy which practical systems should provide. The change in ignition energy requirements with the rate of transfer of energy to the propellant must be recognized in practical ignition system designs. The development of ignition systems with negative temperature coefficients is needed to counterbalance the increase in ignition energies of propellants with a decrease in temperatures. Thermal properties of propellants may be quite important in determining ignition energies. Further studies of these properties may have considerable practical value.

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TABLE I
OVERALL ENERGY TRANSFER TO SUSPENDED COILS
OF COPPER, GLASS, AND PROPELLANT

Number of Coils	Coil Material	Gas	Energy Transferred (cal.)			Energy Transferred per unit area (cal/cm ²)	
			Total System	Blank	Walls* (Corrected blank)	Walls	Coils
1	Glass	Helium	30.8	20.3	17.8	.110	.116
1	Copper	Helium	29.7	20.3	18.0	.111	.118
1	Glass	Argon	15.3	7.3	6.7	.040	.077
1	Copper	Argon	14.2	7.3	6.8	.041	.074
2	Glass	Helium	28.5	19.9	18.2	.112	.066
2	Propellant	Helium	30.2	19.9	17.6	.109	.079
2	Glass	Argon	13.0	6.8	6.4	.038	.042
2	Propellant	Argon	13.6	6.8	6.4	.038	.045

* No corrections were made for competition between walls and coils for heat. Tabulated values are thus higher than actual values for the walls and lower than actual values for the coils.

** Although the tabulated values for the double coils would be expected to be lower than for the single coils, due to the cause noted above, part of the difference in values between single and double coils is real. This is due to the greater inter-coil competition for heat in the double coil experiments.

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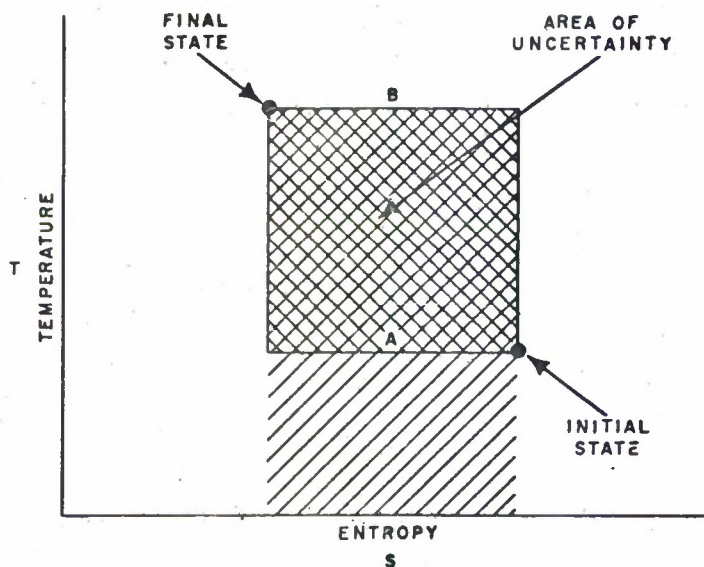
TABLE II
IGNITION ENERGIES* OF PROPELLANTS

Propellant	Ignition Energies cal/cm ²			
	Helium	Argon	Average	110% of Average
Arcite - 141	.117	.116	.117	.129
JPN	.239	.202	.221	.243
IHBE-152	.278	.202	.240	.264
IX-94	.285	.200	.243	.267
Arcite-178	.270	.254	.262	.288
SPDN-9836	.285	.224	.255	.281
N-4	.329	.252	.291	.320
N-5	.296	.270	.283	.311
IMR-6962	.288	.266	.277	.305
OGK	.370	.286	.328	.361
SPCG-7833	.358	.268	.313	.344
EX-7077	.350	>.299	>.325	>.356

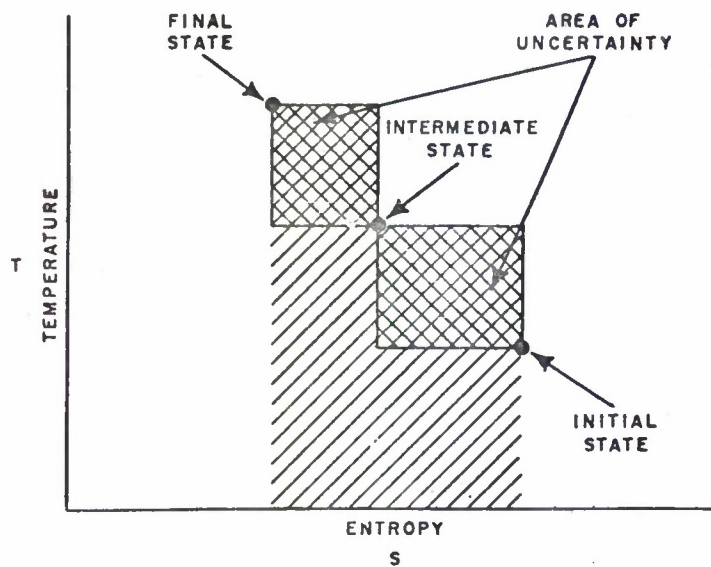
* Ignition energies are entered to 3 sig. figs. for greater accuracy in statistical treatment, but individually 2 sig. figs. would be better.

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(a) INITIAL AND FINAL STATES KNOWN



(b) INITIAL, ONE INTERMEDIATE, AND FINAL STATES KNOWN

FIG.1 HEAT LOSSES DURING A COMPRESSION

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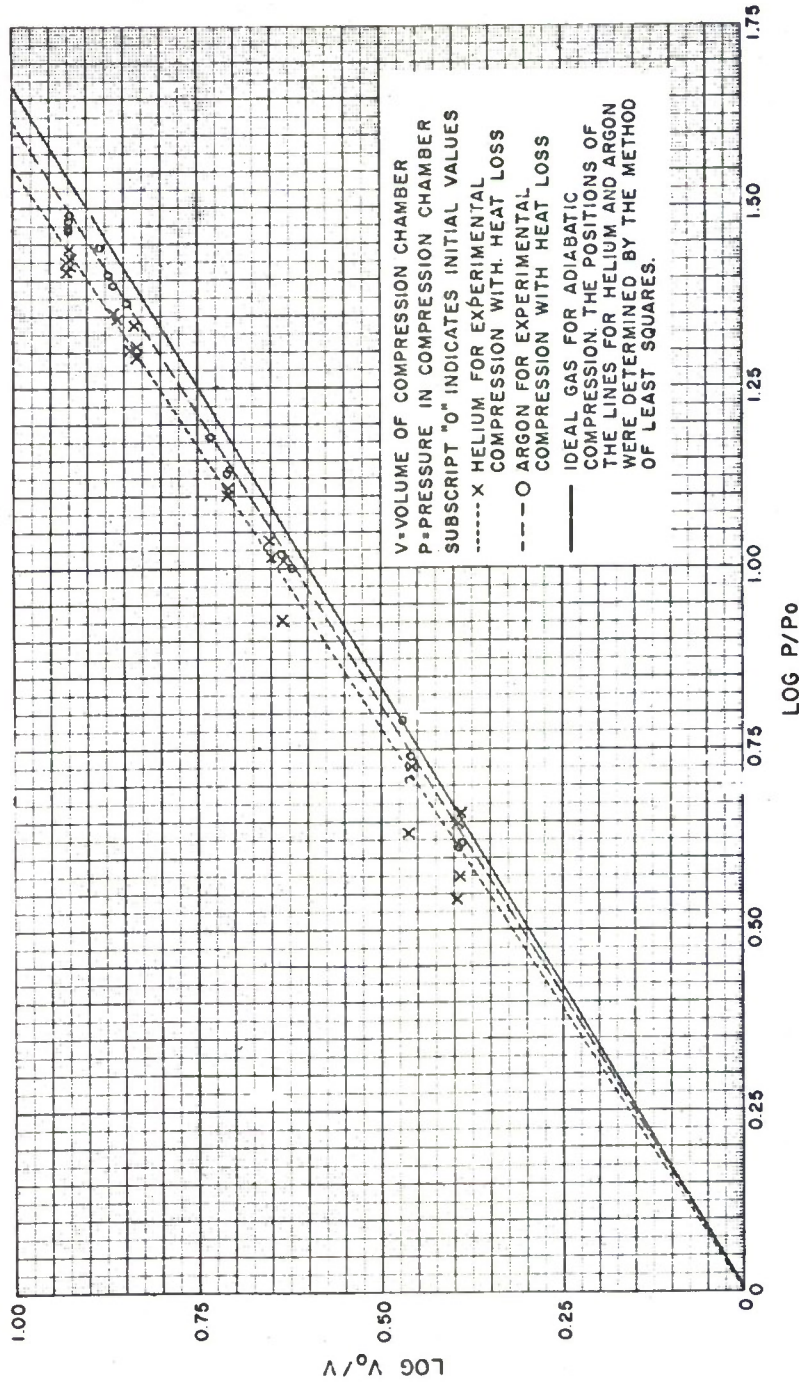


FIG.2 LOG V₀/V VS LOG P/P₀

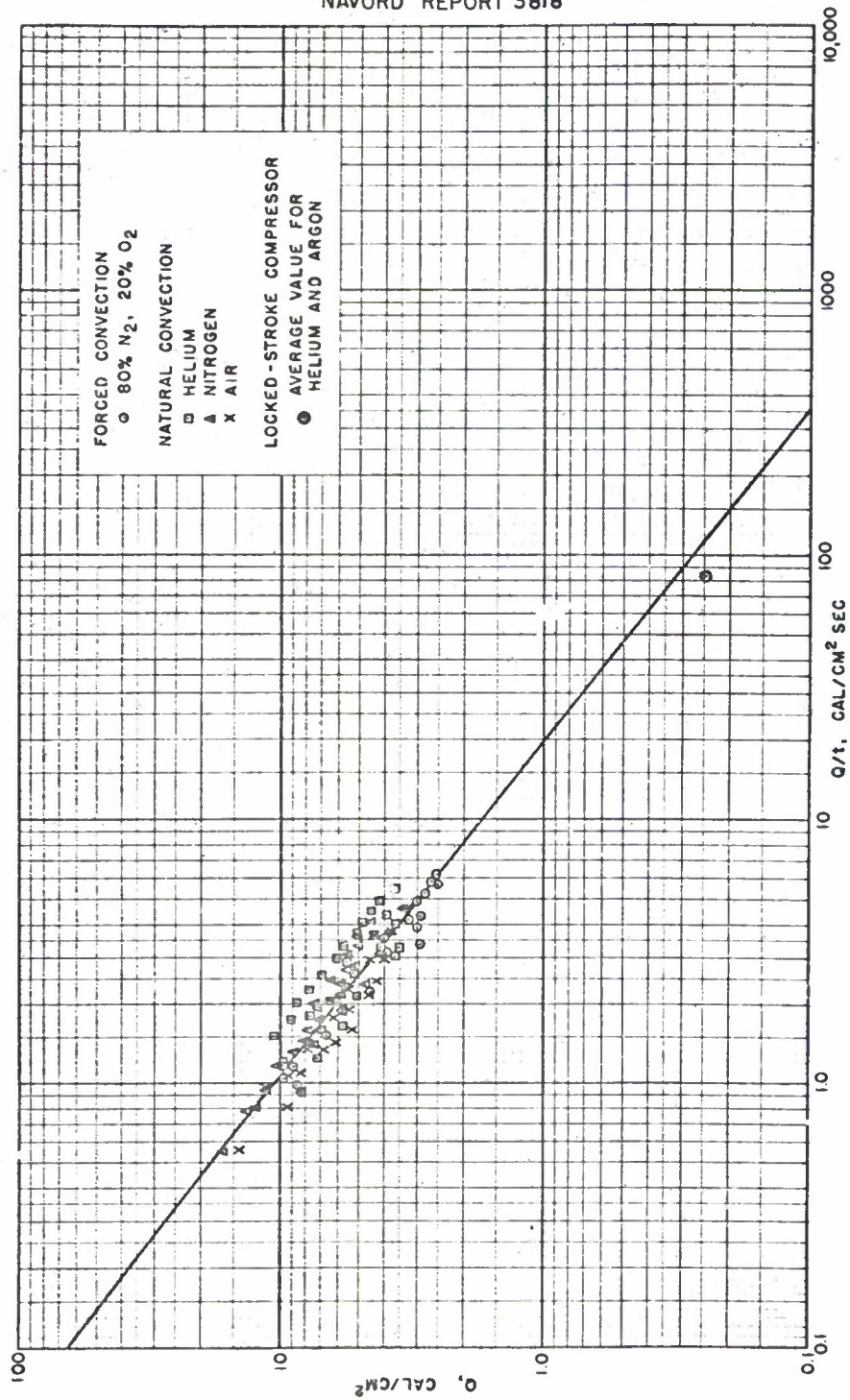


FIG. 3 IGNITION ENERGY VS ENERGY FLUX FOR M-2 PROPELLANT

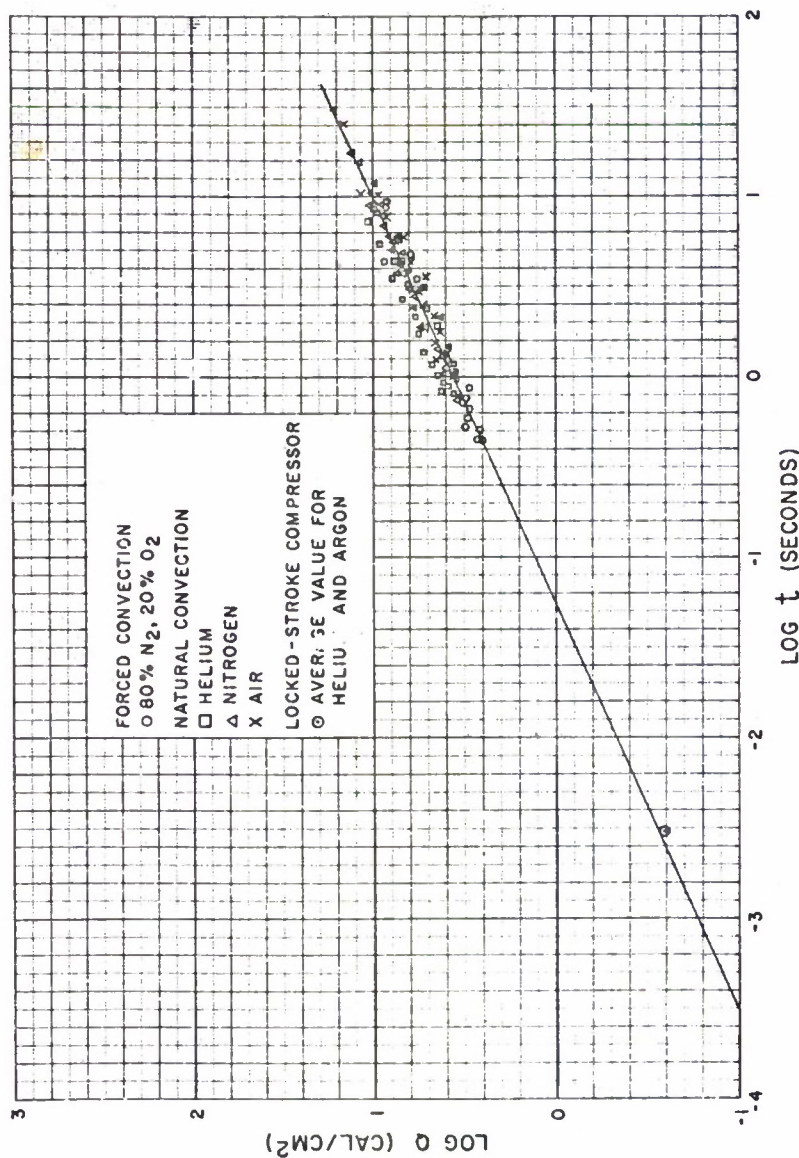


FIG. 4 IGNITION ENERGY VS TIME REQUIRED FOR IGNITION
OF M-2 PROPELLANT

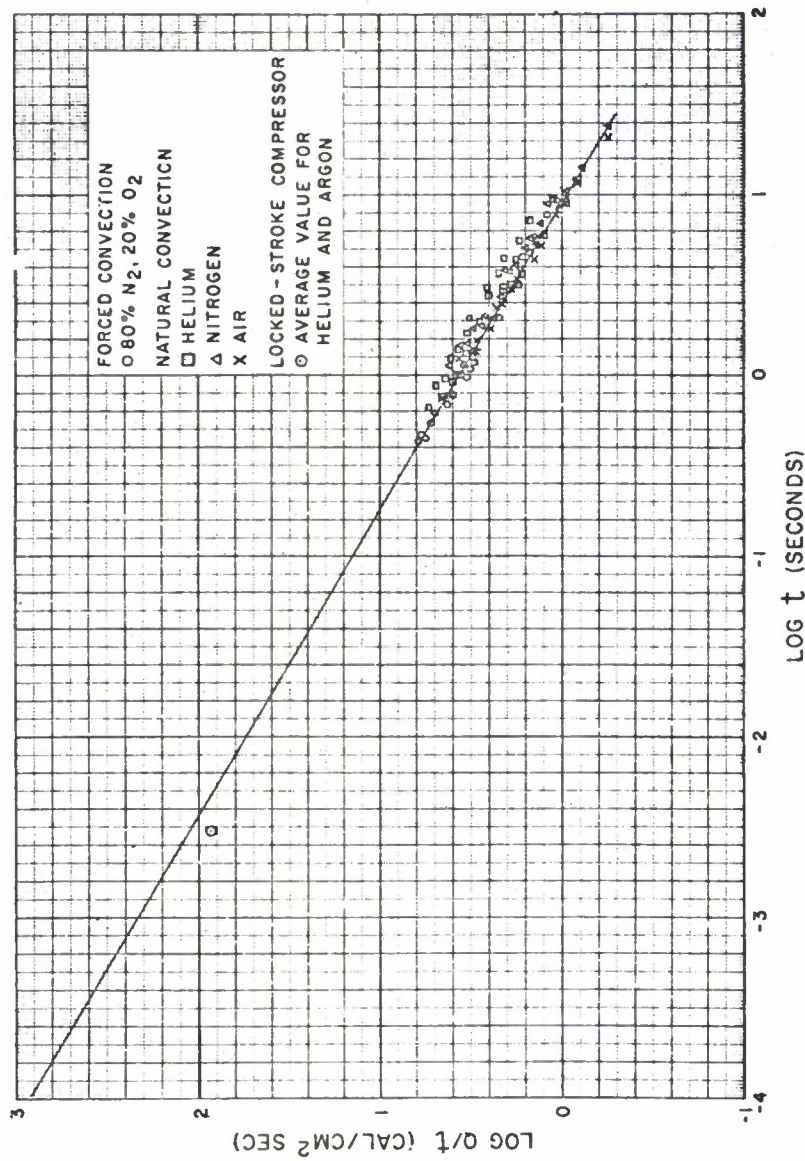


FIG. 5 ENERGY FLUX VS TIME REQUIRED FOR IGNITION
OF M-2 PROPELLANT

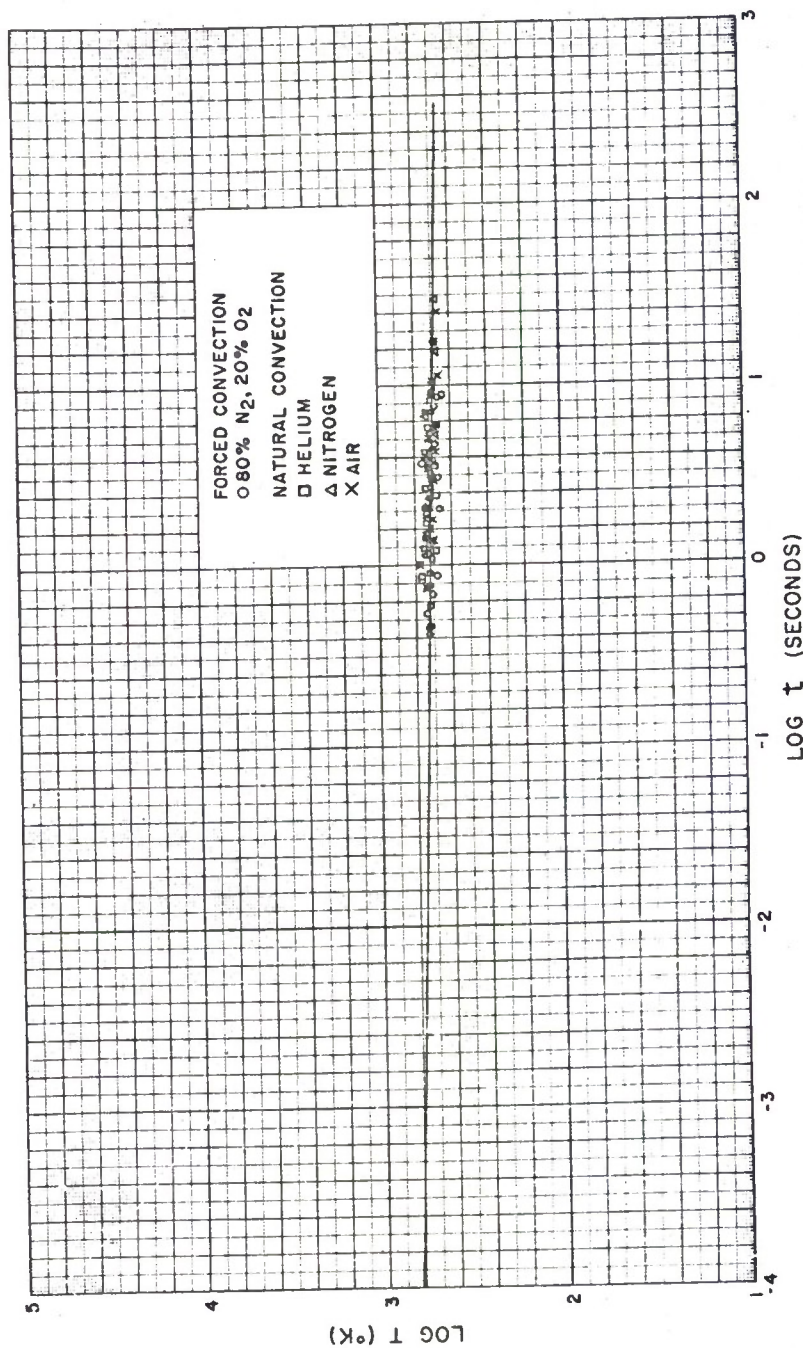


FIG. 6 IGNITION TEMPERATURE VS TIME REQUIRED FOR IGNITION
OF M-2 PROPELLANT

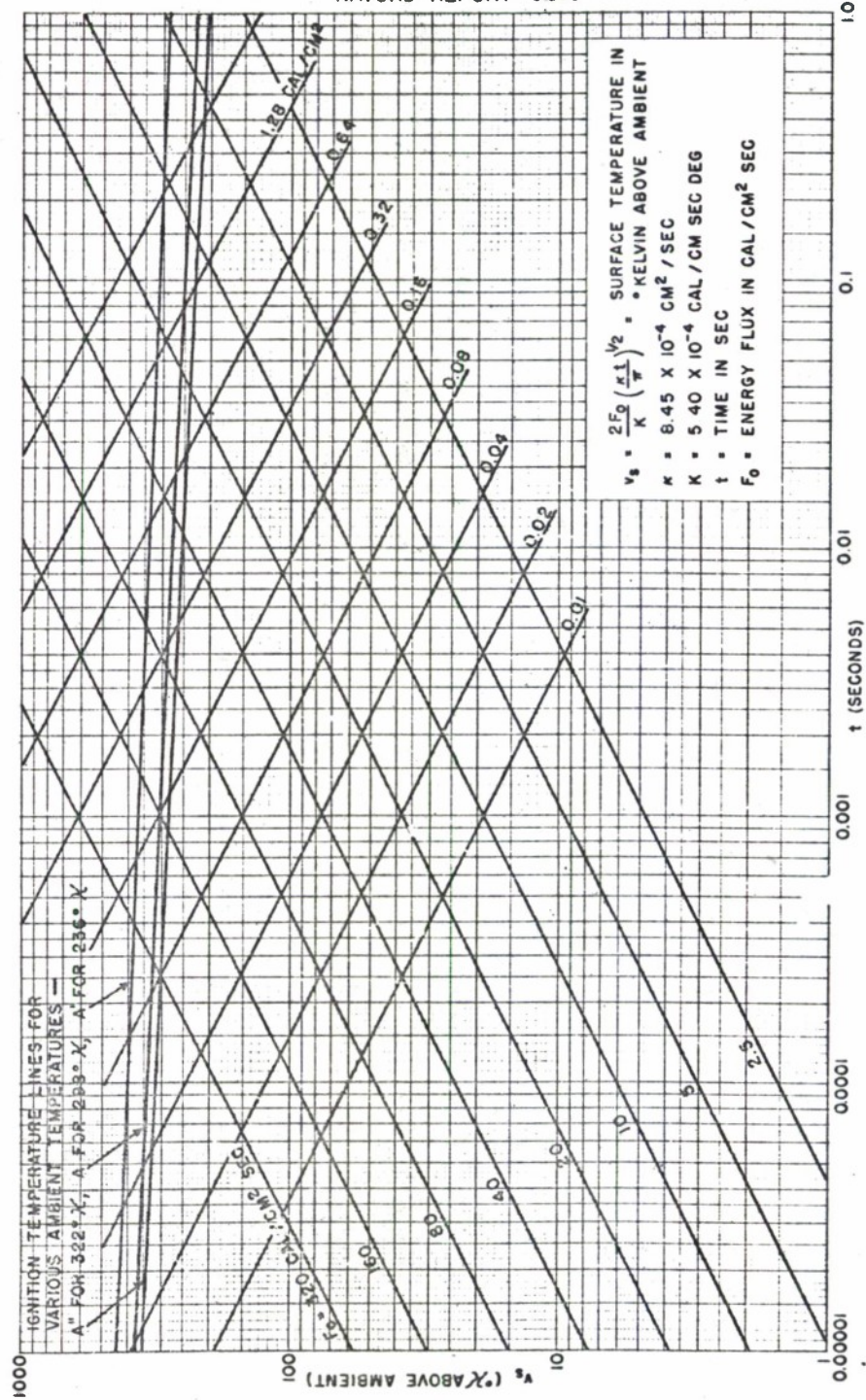


FIG. 7 GRAPHIC MODEL (FOR IGNITION ENERGY, IGNITION TIME, SURFACE TEMPERATURE, AND ENERGY FLUX CALCULATIONS)

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